

IN THE UNITED STATES PATENT AND TRADE MARK OFFICE

In re PATENT APPLICATION of:

Kazuo TAKAOKI

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For: MODIFIED PARTICLE, CATALYST COMPONENT FOR ADDITION POLYMERIZATION, CATALYST FOR ADDITION POLYMERIZATION AND PROCESS FOR PRODUCING ADDITION POLYMER

DECLARATION UNDER 37 C.F.R. 1.132

> Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

I, Kazuo Takaoki, a citizen of Japan, residing at 1-9-324, Yusyudainishi, Ichihara-shi, Chiba, Japan, do hereby declare and say that:

1. I am a member of a team which has been researching and developing a polymerization catalyst, and therefore I am familiar with the subject matter disclosed in the above-identified application;

2. I was graduated from Tokyo University in 1996, and since then, I have been employed by Sumitomo Chemical Company, Limited, where I have been engaged in the research and development works on a polymerization catalyst at the Petrochemicals Research Laboratory of said company;

3. I have read and sufficiently understood the Office Action with the mailing date of May 9, 2006; and

4. In order to attain the purpose as below, the following Comparative Experiment 1 was conducted.

## 1. Purpose

It is a purpose to show unobviousness of the present invention over the cited documents (U.S. 2002/0143124A1 and DE 10164188A1), wherein both documents constitute a family.

## 2. Comparative Experiment 1 and Result

In order to show unobviousness of the present invention over the cited documents (U.S. 2002/0143124A1 and DE 10164188A1):

(1) the following Comparative Experiment 1 was carried out using a combination of diethylzinc ( $(C_2H_5)_2Zn$ ), pentafluorophenol ( $C_6F_5OH$ ) and silica ( $SiO_2$ ), which combination is disclosed in the above-mentioned U.S. 2002/0143124A1, Example 1, columns [0232] and [0234]; and

(2) its result was compared with that of Example 2 of the present application, wherein a combination of triphenyl bismuth ( $Bi(C_6H_5)_3$ ), pentafluorophenol ( $C_6F_5OH$ ) and silica ( $SiO_2$ ) was used.

### Comparative Experiment 1

Comparative Experiment 1 was carried out according to substantially the same method as that mentioned in Example 2 of the present invention, except that triphenyl bismuth ( $Bi(C_6H_5)_3$ ) used in Example 2 was changed to diethylzinc ( $(C_2H_5)_2Zn$ ).

#### (1) Reaction between diethylzinc ( $(C_2H_5)_2Zn$ ) and pentafluorophenol ( $C_6F_5OH$ )

Into a 200 mL four-necked flask purged with nitrogen gas, 10 mL of a hexane solution of diethylzinc ( $(C_2H_5)_2Zn$ ) having a concentration of 1.0 mol/L-solution (therefore, 10 mL of said solution containing 10  $\mu$ mol of  $(C_2H_5)_2Zn$ ) and 50 mL of hexane were introduced, and the resultant mixture was cooled to -78°C. To the mixture, there was added dropwise 20 mL of a hexane solution of pentafluorophenol ( $C_6F_5OH$ ) having a concentration of 1.0 mol/L-solution (therefore, 20 mL of said solution containing 20  $\mu$ mol of  $C_6F_5OH$ ). After completion of dropping, temperature of the resultant mixture was raised to room temperature, and said mixture was stirred for four hours, thereby forming a white solid. Said white solid was separated by filtration, and dried under reduced

pressure, thereby obtaining 3.41 g of a white solid.

(2) Reaction between said white solid and silica ( $\text{SiO}_2$ )

Into a 50 mL four-necked flask purged with nitrogen gas, 0.94 g (2.18 mmol) of the above-obtained white solid and 30 mL of toluene were introduced, and the resultant mixture was heated to 75°C. To the mixture, there was added 0.87 g of silica ( $\text{SiO}_2$ ) having a trade name of SYLOPOL 948, manufactured by Davison Co., Ltd., which had been heat-treated at 300°C. The mixture was stirred for eight hours at 75°C, thereby forming a solid product. Said solid product was washed four times with each 20 mL of toluene, and then once with 20 mL of hexane. The washed solid product was dried under reduced pressure, thereby obtaining 1.23 g of white powder. Said white powder was found by elemental analysis to contain 6.7% by weight of a zirconium atom and 16% by weight of a fluorine atom.

(3) Polymerization

Into a 400 mL-inner volume autoclave equipped with a stirrer, which had been dried under reduced pressure and then purged with argon gas, there were introduced 190 mL of hexane and 10 mL of 1-hexene. The resultant mixture was heated to 70°C. Ethylene was added thereto while keeping its partial pressure of 6 kg/cm<sup>3</sup>. After the system was stabilized, there were added thereto 0.25 mL of a heptane solution of triisobutylaluminum having a concentration of 1  $\mu\text{mol}/\text{mL}$ -solution, 0.5 mL of a toluene solution of ethylenebis(indenyl)zirconium dichloride having a concentration of 2  $\mu\text{mol}/\text{mL}$ -solution (therefore, 0.5 mL of said solution containing 1  $\mu\text{mol}$  of ethylenebis(indenyl)zirconium dichloride), and 30.2 mg of the above-obtained white powder as a solid catalyst component, in this order. Polymerization was carried out for 30 minutes at 70°C while feeding ethylene so as to keep a constant total pressure, thereby obtaining 4.33 g of an ethylene/1-hexene copolymer. Polymerization activity was found to be 290 g/g-solid catalyst component/hour, and 8.7 kg/mmol-Zr/hour. Said copolymer was found to have a weight average molecular weight ( $M_w$ ) of 210,000, a molecular weight distribution ( $M_w/M_n$ ) of 3.7, an SCB value (short chain branch number per 1,000 carbon atoms) of 14.94, and two melting points of 101.4°C and 107.7°C.

### 3. Discussion and Conclusion

The above Comparative Experiment 1 is summarized in the following Table A together with Example 2 of the present invention.

While an object of the present invention is to provide a modified particle which can form a catalyst for addition polymerization exhibiting a high polymerization activity (see the present specification, page 3), Table A shows that:

- Comparative Experiment 1 using a combination " $(C_2H_5)_2Zn + C_6F_5OH + SiO_2$ " gives a polymerization activity of "290 g/g-solid catalyst component/hour" or "8.7 kg/mmol-Zr/hour"; and

- Example 2 of the present invention using a combination " $Bi(C_6H_5)_3 + C_6F_5OH + SiO_2$ " gives a polymerization activity of "1,500 g/g-solid catalyst component/hour" or "45 kg/mmol-Zr/hour"; and therefore, the former polymerization activity is much smaller than the latter one.

Accordingly, I do not think that the present invention is obvious over the cited documents (U.S. 2002/0143124A1 and DE 10164188A1).

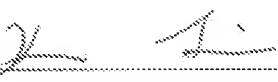
Table A

	Comparative Experiment 1	Example 2
Combination of components	$(C_{2H_5})_2Zn + C_6F_5OH + SiO_2$	$Bi(C_6H_5)_3 + C_6F_5OH + SiO_2$
Polymerization activity		
- g/g-solid catalyst component/hour	290	1,500 (Note)
- kg/mol-Zr/hour	8.7	45
Ethylen/1-hexene copolymer		
- $M_w$	210,000	58,000
- $M_w/M_n$	3.7	1.9
- SCB	14.94	36.0

Note: "1,500 g/g-solid catalyst component/hour" in Table A has the same meaning as that of "1,500 g/g-carrier/h" in Example 2.

I hereby declare further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 25<sup>th</sup> day of August, 2006

  
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Kazuo Takacki